
STEAM BOILERS, POWER-GENERATING FUEL, BURNERS, AND BOILER AUXILIARY EQUIPMENT

Studying the Conversion of Coals and Sludges in Superheated Steam Flow

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Abstract—Results from work on experimentally studying and numerically simulating gasification of low-grade coals and wastes generated from the coal-mining industry by means of steam conversion are presented. The mass concentrations of the gas phase components H₂ and CO obtained at different values of the steam flowrate-to-initial material feed ratio are determined from the calculation results.

Keywords: steam-assisted gasification, wastes from coal production and coal cleaning, numerical simulation, gasification kinetics

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In Russia's new Energy Program, the Government of the Russian Federation has put forward an objective according to which the fraction of coal in the country's energy balance must be increased by approximately a factor of 1.5 by 2020 [1]. The growth of production rates and increase in the coal cleaning fraction will entail a growth of wastes from the coal-mining industry containing an organic component. The amounts of this component are sufficient for their commercial-grade use; however, this component is not quite suitable for processing with the use of existing traditional technologies. High-ash sludges from coal-cleaning factories, culm, and sifting generated during a coal mining process, as well as low-grade coals, can be related to the category of such wastes. On February 27, 2013, the Ministry of the Russian Federation for Energy held a meeting of the Scientific-Technical Council of the Coal Industry, which was devoted to the concept of the project called "Development of a Standard Energy Technology Complex for Integrated Processing of Low-Grade Coals and Wastes from the Coal-Mining Industry with Producing Engine Fuels and Generating Power and Heat." According to this concept, development of technologies for nonfuel use of the organic part of low-grade coals and wastes from the coal-mining industry is becoming of much significance. Gasification by means of superheated steam is one of technologies for processing such materials having high contents of moisture and ash [2, 3]. The advantage of using steam as a gasifying agent is also confirmed by recent foreign studies, the results of which are reported, e.g., in [4–8]. Development of an industry-grade processing technology entails the need to carry out a large

amount of experimental investigations. However, the necessary scope of such investigations can be reduced considerably by combining model experimental studies with numerical simulation of the conversion process. The study results presented in this work are aimed at implementing a technology for subjecting wastes from the coal-mining industry to conversion by means of superheated steam to obtain medium-calorific fuel gas or synthesis gas for the chemical industry, e.g., for producing synthetic liquid fuels.

The experimental investigations of steam-assisted gasification of coal containing wastes were carried out on an experimental setup the design of which is schematically shown in Fig. 1. Samples of coal wastes were subjected to conversion in a dense layer of particles in the medium of steam superheated to a temperature of up to 1200°C without access of oxygen at a pressure slightly higher than the atmospheric pressure. Superheated steam was obtained by firing a stoichiometric hydrogen–oxygen mixture in steam flow [3]. The flowrates of steam and combustible mixture were equal to 5–10 l/min ($t = 120$ – 130°C) and 2–5 l/min, respectively. Coal wastes (culm) from the Bere-zovskaya coal mine with moisture content $W^a = 1.08\%$, content of volatiles $V^d = 19.27\%$, ash content $A^d = 9.59\%$, and elemental composition $N = 1.51\%$, $C = 77.27\%$, and $H = 3.94\%$ were used as initial material. The material was preliminarily subjected to carbonization (during which the volatiles were removed) by holding it at a temperature of 600°C for 30 min. The fraction composition of particles varied in the range 3–4 mm, and the mass of a sample was around 2.5 g. The concentrations of reaction products (CO,

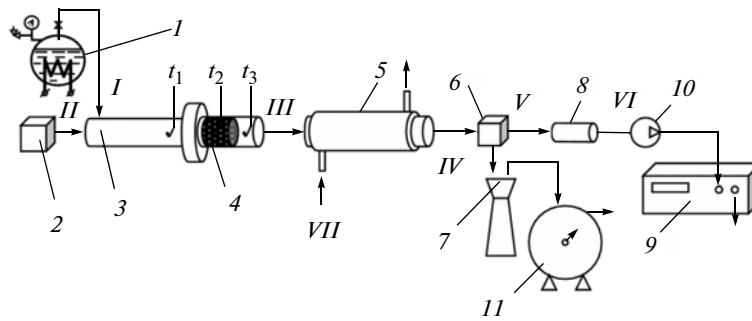


Fig. 1. Schematic design of the experimental setup. (I) Inlet of low-temperature steam, (II) inlet of high-temperature steam, (III) outlet of steam-gas mixture, (IV) outlet of condensate, (V) outlet of wet gas, (VI) outlet of dry gas, and (VII) cooling water. (1) Steam generator, (2) generator of combustible mixture, (3) working section mixing zone, (4) working section reaction zone, (5) condenser, (6) separation reservoir, (7) condensate collection reservoir, (8) gas drying reservoir, (9) gas analyzer, (10) peristaltic pump, and (11) gas drum counter. T_1 , T_2 , T_3 are temperature measurements upstream of the reaction zone, in this zone, and downstream of it.

H_2 , CO_2 , O_2 , and CH_4) were measured using a Test-1 flow-type gas analyzer.

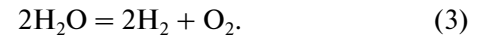
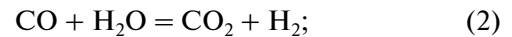
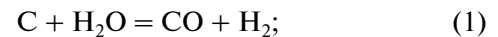
Dependences characterizing the formation of the principal products from the steam-assisted gasification process as functions of its duration and steam temperature at constant steam flowrates were obtained as a result of the experimental investigations. The loss of sample mass as a result of conversion during the experiment was from 12% at 735°C to 58% at 1000°C.

Figure 2 shows the typical results obtained from measurements of temperature in a sample and volume concentrations of components in the obtained mixture of gases. In the time interval 0–300 s, the sample is heated, and the air contained in it is displaced. A noticeable concentration of O_2 in the gasification products can be attributed to catalytic reduction of CO_2 on the ash frame, as was described in [9].

The results obtained from sample temperature measurements were used for determining the coefficient of heat transfer from steam flow to granulated carbon raw material during an abrupt increase of heat carrier temperature at the layer inlet according to the procedure described in [10–12]. For calculating the heat transfer coefficient according to this procedure, we used an analytical approximation of the time dependence of the granulated medium's thin layer temperature at the initial stage of the heating process. The thermophysical properties were taken to be constant. It was assumed that in heating the grains of medium, the temperature field remains uniform in the grain volume. The results of temperature measurements were subjected to processing, and it was found from that processing that the heat-transfer coefficient is described with good accuracy by the following formula: $Nu = 0.03772Re^{0.8}$ [13].

The experiments were carried out under the conditions of a large excess of H_2O and with small concentrations of the remaining components. In view of these circumstances, the following reactions were taken into

account for approximately describing the kinetics of steam conversion:



These reactions are stoichiometric ones; i.e., they characterize the correlation between the amounts of reagents coming in the reaction and the products generated during the carbon gasification process. The presented sequence of reactions does not correspond to their actual sequence.

For obtaining quantitative data on the reaction rates, a technique for processing experimental data was developed. It was assumed in its development that

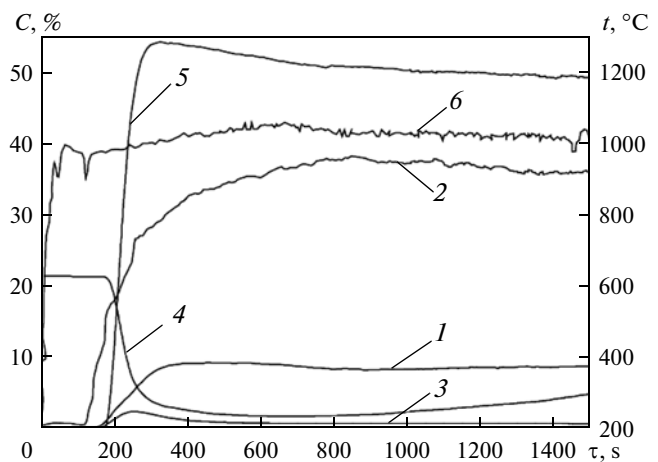


Fig. 2. Dependences characterizing the generation of characteristic reaction products during gasification of coal wastes from the Berezovskaya mine (under stationary conditions). The temperature in the material bed is 1000°C. Volume concentrations: (1) CO_2 , (2) CO , (3) CH_4 , (4) O_2 , and (5) H_2 . (6) Temperature in the bed t_2 .

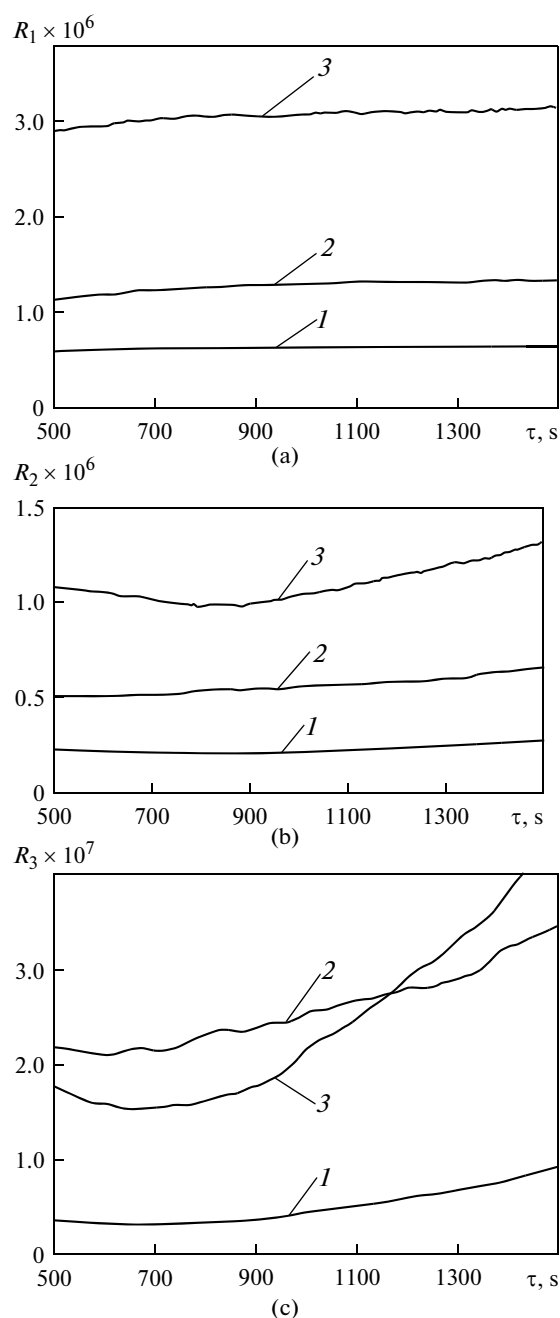


Fig. 3. Time dependences of mass rate for reactions (1)–(3) (a–c, respectively) for the carbonized coal wastes from the Berezovskaya mine. Process temperature, °C: (1) 735, (2) 880, and (3) 1000.

the volume yield of conversion products remains constant. For determining the mass yield of components, we calculated the relative mass yield of carbon M_C in these components by numerically integrating the products of experimentally obtained volume concentrations of CO and CO₂ by the densities of these components and their volume flowrate (an unknown quantity). Knowing the ratio between the change in the sample mass ΔM_C and the relative yield of carbon

M_C , it is possible to determine the norm of the time dependence of carbon yield $\Delta m_C(\tau)$, kg/s, as well as the absolute time dependences of mass yield $\Delta m_{O_2}(\tau)$, $\Delta m_{CO}(\tau)$, and $\Delta m_{CO_2}(\tau)$, kg/s.

We denote the mass rates of reactions (1)–(3) by ΔM_j , kg/s, where j is the reaction number. Then, for the system of reactions we obtain the following system of linear equations for the resulting component yield rates:

$$\frac{\mu_{CO}}{\mu_{CO} + \mu_{H_2}} \Delta M_1 - \frac{\mu_{CO}}{\mu_{CO} + \mu_{H_2O}} \Delta M_2 = \Delta m_{CO}(\tau);$$

$$\frac{\mu_{CO_2}}{\mu_{CO_2} + \mu_{H_2}} \Delta M_2 - \Delta M_3 = \Delta m_{CO_2}(\tau);$$

$$\frac{\mu_{O_2}}{\mu_C + \mu_{O_2}} \Delta M_3 = \Delta m_{O_2}(\tau).$$

From this system of linear equations, the mass rates of reactions (1)–(3) are calculated for each measurement using the known dependences for the yield of components. For verifying the procedure, we determined the mass yield of H₂ from the calculated rates of reactions (1)–(3) using the following formula:

$$\Delta m_{H_2} = \frac{\mu_{H_2}}{\mu_{CO} + \mu_{H_2}} \Delta M_1 + \frac{\mu_{H_2}}{\mu_{CO_2} + \mu_{H_2}} \Delta M_2.$$

The largest mismatch between the typical calculation results and experimental data does not exceed 15%, which testifies the validity of the applied procedure. Figure 3 shows the graphs of mass velocities R_1 – R_3 obtained from processing the experimental data for carbonized wastes of coal from the Berezovskaya mine.

In the time interval 500–1500 s, the rates of reactions (1) and (2) are almost constant and increase monotonically with temperature. The rate of catalytic reaction (3) in this interval of time varies with temperature in nonmonotonic manner, which can be attributed to a heterogeneous composition of the ash frame through which steam is seeping. A noticeable growth in the rate of reaction (3) with temperature is caused by an increase in the thickness of ash frame with decreasing the sample mass.

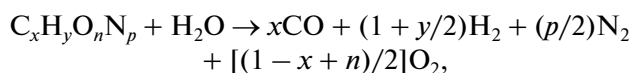
Numerical simulation of the gasification process was carried out for a gasifier made in the form of a vertical cylindrical thermally insulated apparatus into which dispersed carbon-containing material is supplied from above. Moving downward under the effect of its own weight (with the ash residue removed from the gasifier lower part), this material forms a porous medium, through which superheated steam moves in a counterflow manner. Preliminary assessments of the intensity of interphase heat transfer showed that the height of the shell must be considerably larger than its diameter. This means that the system under study can be modeled in 1D approximation with ignoring the

nonuniformity of the main parameters (velocity and temperature) over the shell section.

The model of autothermal bed gasification described in [14] was laid as a basis in developing the model. Under the conditions of autothermal steam–air gasification, the endothermic reaction of interaction between carbon and H_2O proceeds as part of carbon burns in oxygen. Unlike steam–air gasification, there is no oxygen during steam-assisted gasification, and the heat required for this reaction to proceed is brought about by superheated steam. Owing to this circumstance, it becomes possible to simplify the model described in [14].

This model assumes that the dispersed phase consists of spherical particles having the same diameter, with a rigid ash skeleton, the pores of which are uniformly filled by carbon, moisture, and volatiles. The substance of particles consists of carbon, moisture, volatiles, and inert ash residue. The steam–coal particles system has the form of a heterogeneous mixture. The dispersed phase moves from the top down under the effect of its own weight, and the gaseous phase is seeping in a countercurrent manner (Fig. 4). In simulating the processes that take place in the solid phase, like pyrolysis and heterogeneous reaction, and in describing the heat conductivity of the solid phase, the continual approach is used [15]. The solid phase temperature at a fixed point is the surface temperature of particles averaged over the apparatus section (temperature and concentration gradients in a particle were not taken into account). The solid phase hydrodynamic and thermophysical properties (its resistance to the seeping gas flow, heat conductivity, and heat capacity) are uniformly distributed over the bed height and do not vary in the radial direction. The gas phase includes superheated steam, gaseous products of pyrolysis, and products of heterogeneous and homogeneous chemical reactions, which exist in the form of a homogeneous mixture of ideal gases obeying the Clapeyron–Mendeleev equation of state. The pressure is taken to be constant and equal to atmospheric pressure.

The thermal decomposition of coal particles is regarded as transition of moisture and volatiles from the solid phase into the gaseous one. The drying and volatile release processes are assumed to obey the Arrhenius dependences of rates on temperature. The volatiles are represented by hypothetical substance $C_xH_yO_nN_p$. For describing the chemical reactions of gas components at the appropriate temperatures, we took into account the rates of homogeneous reactions proceeding according to the following mechanism [14]:



where x , y , p , and n are the molar concentrations of elements in the volatiles, which have been determined from the data of elemental analysis of coal and are taken to be constant.

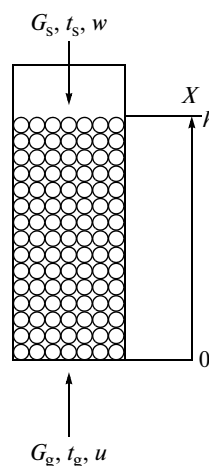


Fig. 4. Calculation scheme.

The system of equations in the model includes the transfer equations for the gas phase components, the gas phase energy equation, the transfer equations for the solid phase components, and the solid phase energy equation. A more detailed description of the model is given in [16].

The organic raw material gasification process in a gasifier with the steam flow thermal capacity equal to 50 kW was studied numerically. With this thermal capacity and temperature of steam $t_g = 1000^\circ\text{C}$, the gasifier cross-section area S is $4.4 \times 10^{-3} \text{ m}^2$, and the shell radius $R = 0.0375 \text{ m}$. The flowrate of steam entering into the gasifier at this capacity $G_g = 0.0189 \text{ kg/s}$. Coal sludge from the Berezovskaya central coal-cleaning factory having the following content (in mass fractions) was selected as solid fuel for modeling the gasification process: moisture $W^r = 35\%$, carbon $C^r = 42.6\%$, hydrogen $H^r = 2.9\%$, oxygen $O^r = 13.9\%$, nitrogen $N^r = 0.4\%$, and volatiles $V^r = 28.8\%$. The calculated molar coefficients for volatiles $C_xH_yO_nN_p$ are as follows: $x = 2.0374$, $y = 6.0695$, $n = 1.8328$, and $p = 0.0602$; the molar mass of volatiles is equal to 0.0608 kg/mol . The solid phase grain diameter $d = 0.004 \text{ m}$, the gasifier height $h = 1 \text{ m}$, and the solid phase initial temperature $t_g = 20^\circ\text{C}$. The solid phase flowrate was selected from the following ratios of flowrates: $K_g = 1, 0.5, 0.25$, and $0.16\text{--}0.176$, where $K_g = G_s/G_g$ is the ratio of the initial solid phase flowrate to the steam flowrate.

Figure 5 shows the profiles of gas and liquid phase temperatures over the working section length. The gas phase temperature decreases from 1000°C at the gasifier inlet to $500\text{--}250^\circ\text{C}$ at its outlet depending on the solid phase flowrate. The profile of solid phase temperature t_s varies in a similar manner. As the solid phase moves on, its temperature t_s increases from the coordinate $x = h$ to the coordinate $x = 0$. The solid phase temperature at the working section outlet varies

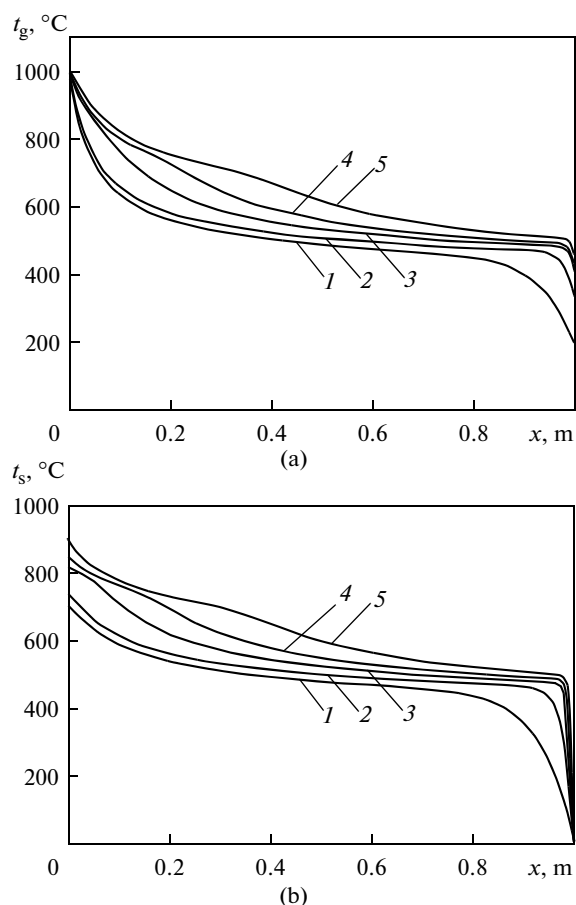


Fig. 5. Temperature profiles of the gas phase t_g (a) and solid phase t_s (b) over the working section height at different values of K_g . K_g values: (1) 1, (2) 0.5, (3) 0.25, (4) 0.2, and (5) 0.16. The solid phase grain diameter is $d = 0.004$ m.

in the range from 700 to 900°C. As the solid phase flowrate decreases, the average gas phase temperature and the solid phase temperature increase. Figure 6 shows the variation in the gas and solid phase flowrates. It can be noticed that a decrease in the solid phase flowrate G_s does not always lead to a decrease in the gas phase flowrate G_g . For example, when the relative flowrate of solid phase K_g decreases from 1 to 0.5, the gas phase flowrate increases. To explain this fact, we will consider the data on relative mass concentrations of volatiles V_s and carbon C_s in the solid phase (Fig. 7). The calculation results showed that the profile of relative mass concentration of moisture W_s in the solid phase varies in a way similar to that of mass concentration of volatiles V_s in this phase. This is attributed to the fact that the release rates of moisture and volatiles are determined in the mathematical model by the same dependence on the solid phase temperature. This is why the zones of drying and carbonization

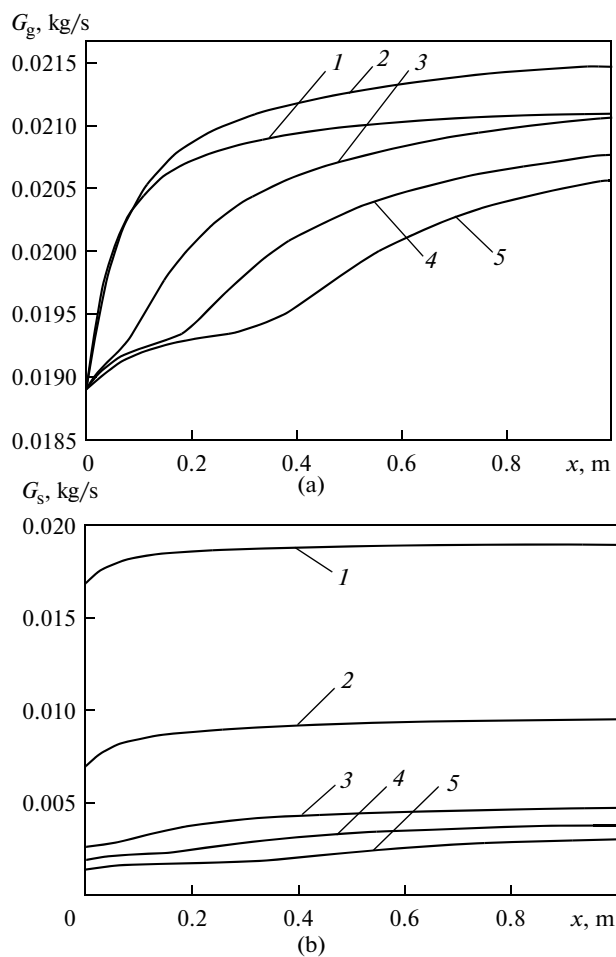


Fig. 6. Mass flowrate profiles of the gas phase G_g (a) and solid phase G_s (b) over the working section height at different values of K_g . The notation is the same as in Fig. 5.

coincide with each other in this model. The full release of moisture W_s and volatiles V_s (Fig. 7a) over the gasifier's entire height takes place when the ratio of flowrates K_g is equal to around 0.25. As the relative solid phase flowrate is decreased further, the drying zone and the volatile release zone shift toward the gasifier lower part. We now consider the profile of carbon concentration C_s shown in Fig. 7b. An essential change in the concentration profile occurs only if the flowrate coefficient K_g is less than 0.25. This is attributed to the fact that the temperature under such conditions is rather low for conversion of carbon due to the release of moisture and volatiles. The limiting flowrate $K_g = 0.16$ was obtained in decreasing the mass concentration of carbon C_s down to 10%. Figure 8 shows the profiles of mass concentrations of hydrogen m_{H_2} and carbon monoxide m_{CO} .

In analyzing the mass concentration profiles of hydrogen m_{H_2} and carbon monoxide m_{CO} in the gas

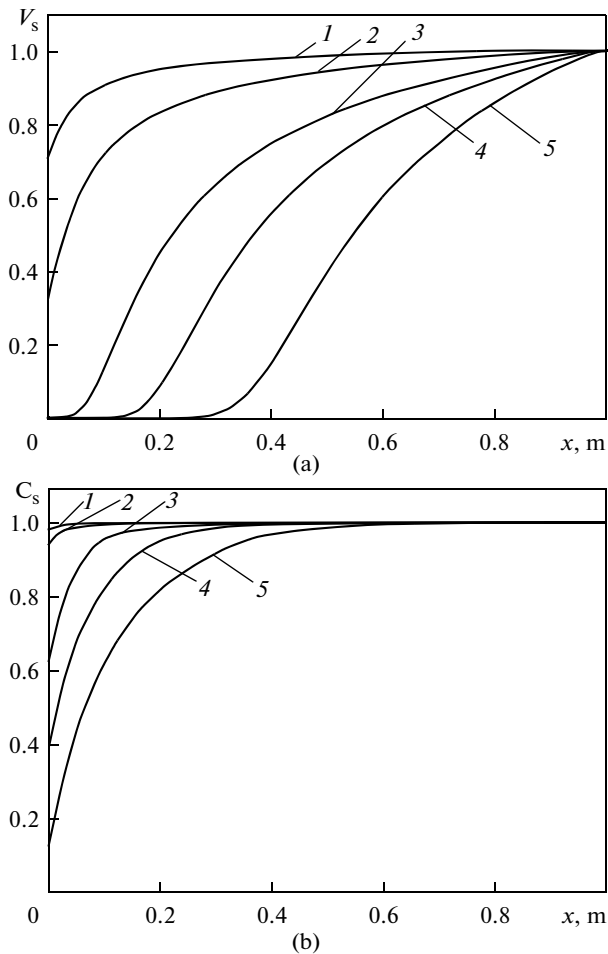


Fig. 7. Profiles of relative mass concentration of volatiles V_s (a) and carbon C_s (b) in the solid phase over the working section height at different values of K_g . The notation is the same as in Fig. 5.

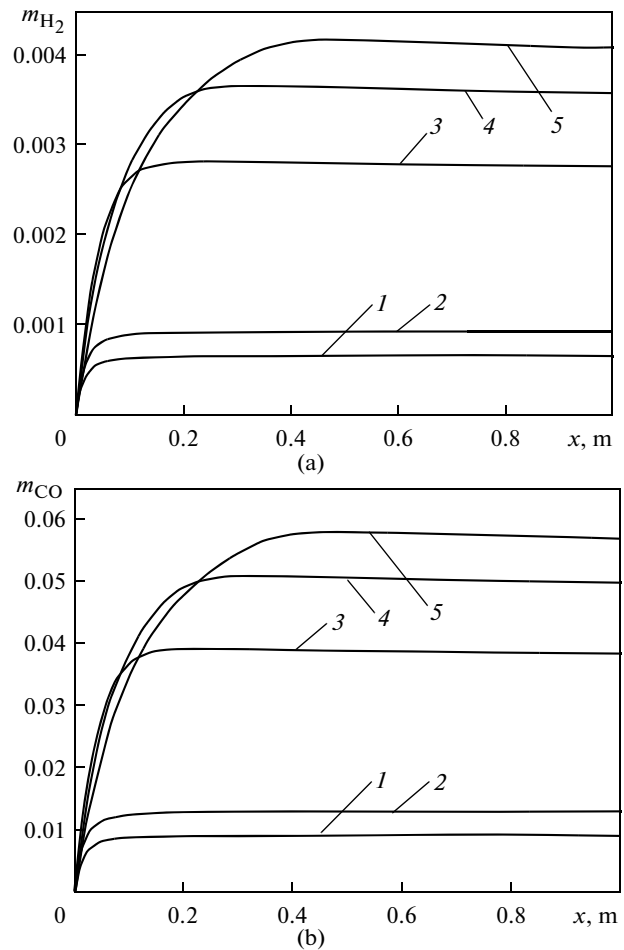


Fig. 8. Profiles of mass concentration of H_2 m_{H_2} (a) and CO m_{CO} (b) in the gas phase over the working section height at different values of K_g . The notation is the same as in Fig. 5.

phase, their dependence on the change of the mass concentration of carbon in the solid phase was found. This points to the fact that the steam-assisted carbon conversion process is a factor playing the principal role in the variation of mass concentration of hydrogen m_{H_2} and carbon monoxide m_{CO} .

An analysis of changes occurring in the profiles of mass concentration of moisture in the solid phase and volatiles obtained in the course of numerical simulation revealed that the drying and carbonization zones (the pyrolysis and volatile oxidation zones) coincide with each other, which is due to the way in which the volatiles and moisture release processes are described in the mathematical model. As the solid phase flowrate is decreased with the initial gas phase flowrate kept constant, the carbon conversion zone increases, and the drying and carbonization zones decrease. The increase in the mass concentration of hydrogen and carbon monoxide in the gas phase is determined by the

steam conversion of carbon in the solid phase. The steam conversion of volatiles in the gas phase is negligibly small.

It can be pointed out in conclusion that in stating the problem we selected the technological scheme for conversion of carbon-containing materials in the form of a single-stage apparatus. The investigation of the characteristics of such apparatus that was carried out in this work revealed that an attempt to realize drying, carbonization, and steam conversion processes in a single apparatus is a contradictory task. In order to achieve better performance characteristics of steam conversion of carbon-containing materials, development of a two-stage technology involving separate drying and carbonization of carbon-containing materials and subjecting the carbonized product obtained at the first stage to steam conversion seems to be a promising approach. Implementation of a numerical experiment for the two-stage technology is a subject of further study.

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